

# Spin-excitation spectra and resistance minima in amorphous ferromagnetic alloys\*

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Resistance minima have been found in recent years to occur in amorphous ferromagnetic alloys below the magnetic ordering temperature. Although a well-developed theory exists for resistance minima in very dilute alloys, the meaning of the phenomena has remained in question for alloys in which the neglect of spin-spin interactions is not justifiable. In this paper it is shown that the observation of resistance minima implies that these alloys have a finite density of near zero frequency excitations. Specifically, the theory of inverse transport coefficients, reformulated in terms of linear response, is used to derive a general expression for the resistivity due to the conduction-electron-spin interaction. Expanding perturbatively, the  $n$ th-order contribution is determined by an  $n$ th-order spin correlation function. To third order it is shown that the coefficient of the  $\ln kT$  term responsible for the resistance anomaly according to the accepted Kondo theory receives contributions in the low-temperature limit only from those parts of the spin correlation functions which have frequencies less than  $kT_K/\hbar$  where  $T_K$  is the Kondo temperature.

## I. INTRODUCTION

The resistance minimum phenomenon observed in metals with dilute concentrations of magnetic impurities has been extensively studied in recent years.<sup>1</sup> The theoretical effort has centered on considering the interaction of the conduction electrons with a single local moment, the "Kondo problem." It has been established that the internal degrees of freedom associated with the possible orientations of the local moment make the calculation of the scattering cross section a many-body problem. The result is a cross section increasing with decreasing temperature, and therefore an increasing resistance at low temperatures.<sup>2</sup> Much of the experimental effort has gone into studying alloys of sufficient diluteness that the conditions of the theorists single-local-moment problem are well approximated.

For alloys with appreciable concentrations of magnetic elements it is clearly not justifiable to neglect the interaction between moments, and the theoretical problem is more complex. It is observed that in crystalline alloys below the magnetic-ordering temperature no resistance minimum is observed, presumably because the individual moment no longer has a choice of orientation. On the other hand, amorphous ferromagnetic alloys continue to exhibit resistance minima well below the magnetic-ordering temperature.<sup>3</sup> Certainly, an important theoretical problem is to develop models for the interaction between spins which will explain these differences.

In this paper we would like to turn the question around by asking what the observation of a resistance minimum indicates about the nature of the spin-excitation spectrum. One way this problem has been addressed is to consider various forms

for magnon dispersion relations and calculate the resistance using the Boltzmann expression.<sup>4</sup> Our approach will be to make use of force-force correlation-function expressions for inverse transport coefficients. These have the advantage, as we shall show below, that the resistivity is related directly to the spin correlation functions whose properties we wish to establish.

In our considerations we will assume that, as in the Kondo theory of dilute alloys, the resistance-minimum phenomenon manifests itself as a  $\ln kT$  term in the third-order contribution to the resistivity due to the conduction-electron-spin interaction. Our calculation will focus on deriving an expression for the coefficient of the  $\ln kT$  term and finding the conditions for it to be nonzero in the low-temperature limit.

In Sec. II we provide a derivation of our expression for the resistivity as an approximation to the linear-response expression for the conductivity. The principal result of Sec. II is Eq. (23). In Sec. III we consider the resistivity in a ferromagnetic material in which the interaction between spins is not negligible. The principal result of Sec. III is Eq. (39), which expresses the relation between the third-order contribution to the resistivity and a triple-spin-correlation function. In Sec. IV the coefficient of the  $\ln kT$  term is singled out and shown to receive contributions only from near-zero-frequency excitations in the low-temperature limit. Section V contains some concluding remarks.

## II. LINEAR-RESPONSE THEORY AND INVERSE TRANSPORT COEFFICIENTS

The theory of linear response as formulated by Kubo provides a presumably exact expression for the conductivity in terms of the current-current

correlation function.<sup>5</sup> As is well known, the application of such expressions has been limited because they give divergent results in any finite order perturbative calculation. In an attempt to avoid these difficulties several authors<sup>6,7</sup> have published, in the recent literature, expressions for inverse transport coefficients such as the resistivity which involve force-force correlation functions. Inverse transport coefficients have the practical advantage that each order in the perturbative expansion is finite.

In this section we wish to obtain a force-force expression for the resistivity in a form more suitable for a perturbative expansion than other expressions in the recent literature.

Our starting point will be the linear-response expressions for the conductivity as formulated by Kubo. The change in the expectation value of a physical quantity  $O(x, t)$  due to an external perturbation  $H_H^{\text{ext}}(t)$  turned on at time  $t = 0$  is

$$\delta\langle O(t) \rangle = \frac{i}{\hbar} \int_0^t dt' e^{-it'} \text{Tr} \{ \rho_G [H_H^{\text{ext}}(t'), O_H(t)] \}, \quad (1)$$

where  $\rho_G$  is the grand canonical statistical operator. For the conductivity we must calculate the change in expectation value of the current operator

$$\begin{aligned} J_\mu(x, t) = & -\sum_\alpha \frac{\hbar}{2mi} \{ [\nabla_\mu \psi_\alpha^\dagger(x, t)] \psi_\alpha(x, t) \\ & - \psi_\alpha^\dagger(x, t) [\nabla_\mu \psi_\alpha(x, t)] \} \\ & - \frac{e}{mc} \psi_\alpha^\dagger(x, t) \psi_\alpha(x, t) A_\mu(x, t), \end{aligned} \quad (2)$$

due to a vector potential  $A_\mu(x, t)$  turned on at time  $t = 0$ :

$$H_H^{\text{ext}}(t) = -\frac{e}{c} \int d^3x J_\mu(x, t) A_\mu(x, t). \quad (3)$$

Then

$$\begin{aligned} \langle J_\mu(x, t) \rangle = & \frac{-i}{\hbar} \int d^3x' \int_0^t dt' \text{Tr} \\ & \times \{ \rho_G [J_\nu(x', t'), J_\mu(x, t)] \} \\ & \times \frac{e}{c} A_\nu(x', t') - \frac{en(x)}{mc} A_\mu(x, t). \end{aligned} \quad (4)$$

We may extend the lower limit of the  $t'$  integral to  $-\infty$  by including the heaviside function  $\Theta(t')$  in the field  $A_\nu(x', t')$ . We also note that

$$J_\mu(x, t) = \sum_\alpha \frac{\partial}{\partial t} [\psi_\alpha^\dagger(x, t) x_\mu \psi_\alpha(x, t)] \quad (5)$$

$$\equiv \frac{\partial}{\partial t} \hat{X}_\mu(x, t). \quad (6)$$

Integrating by parts in the variable  $t'$ , we find

$$\langle J_\mu(x, t) \rangle = + \frac{i}{\hbar} \int d^3x' \int_{-\infty}^{\infty} dt' \theta(t-t')$$

$$\begin{aligned} & \times \text{Tr} \{ \rho_G [\hat{X}_\nu(x', t'), J_\mu(x, t)] \} \\ & \times \frac{e}{c} \frac{\partial}{\partial t'} A_\nu(x', t'), \end{aligned} \quad (7)$$

where we have used the relation

$$(i/\hbar) [\hat{X}_\nu(x', t), J_\mu(x, t)] = [-n(x)/m] \delta(x-x'). \quad (8)$$

The electric field is related to the vector potential by

$$E_\nu(x, t) = -\frac{1}{c} \frac{\partial}{\partial t} A_\nu(x, t). \quad (9)$$

The average current density is

$$\begin{aligned} \langle J_\mu(t) \rangle = & \frac{1}{\Omega} \int d^3x J_\mu(x, t) \\ = & \int \frac{d\omega}{2\pi} e^{-i\omega t} \langle J_\mu(\omega) \rangle, \end{aligned} \quad (10)$$

where  $\Omega$  is the volume of the solid. The frequency-dependent conductivity  $\sigma_{\mu\nu}(\omega)$  is defined by

$$e\langle J_\mu(\omega) \rangle \equiv \sigma_{\mu\nu}(\omega) E_\nu(\omega). \quad (11)$$

We therefore find

$$\begin{aligned} \sigma_{\mu\nu}(\omega) = & \frac{-ie^2}{\hbar\Omega} \int d^3x d^3x' d(t-t') e^{i\omega(t-t')} \Theta(t-t') \\ & \times \text{Tr} \{ \rho_G [\hat{X}_\nu(x', t'), J_\mu(x, t)] \}, \end{aligned} \quad (12)$$

which is merely a variant on the more commonly quoted Kubo expression.

Hence, we see that the central problem in the calculation of the transport is the determination of a function  $I_{\mu\nu}(t-t')$  which is defined to be

$$\begin{aligned} I_{\mu\nu}(t-t') = & \frac{-i\theta(t-t')}{\hbar\omega} \int d^3x d^3x' \\ & \times \text{Tr} \{ \rho_G [\hat{X}_\nu(x', t'), J_\mu(x, t)] \}. \end{aligned} \quad (13)$$

For convenience, we will assume that the matrix  $I_{\mu\nu}(t-t')$  can be diagonalized by a time-independent coordinate transformation; this assumption is surely valid for isotropic, cubic, and cylindrically symmetric problems such as will be dealt with here. In the rest of the paper we will work in the coordinate system in which  $I$  is diagonal and label the diagonal elements  $I_\mu$ .

Kubo has discussed various methods of approximating  $I_\mu(t)$ .<sup>8</sup> For the purpose of computing the dc conductivity it is convenient to use an approximation that would produce a frequency-dependent conductivity of the Drude form. That is, we assume that

$$I_\mu(t) = (n/m) e^{-t/\tau_\mu} \theta(t). \quad (14)$$

This expression for  $I$  yields a frequency-dependent conductivity of the form

$$\sigma_\mu(\omega) = ne^2\tau_\mu/m(1-i\omega\tau_\mu); \quad (15)$$

and the dc conductivity and resistivity are given by

$$\sigma_{\mu}(0) = (ne^2/m)\tau_{\mu} \quad (16)$$

and

$$R_{\mu} = m/ne^2\tau_{\mu}, \quad (17)$$

respectively. Kubo<sup>8</sup> suggests an approximate procedure for evaluating  $\tau_{\mu}$ . That is, one takes

$$\frac{1}{\tau_{\mu}} = \lim_{\eta \rightarrow 0^+} \frac{m}{n} \int_{\eta}^{\infty} \frac{\partial^2}{\partial t^2} I_{\mu}(t) dt. \quad (18)$$

Hence, substituting Eqs. (13) into (18) and (18) into (17), we obtain

$$R_{\mu} = \lim_{\eta \rightarrow 0^+} \frac{m^2}{n^2 e^2 \Omega} \frac{i}{\hbar} \int d^3x d^3x' \times \int_{\eta}^{\infty} dt \text{Tr} \left[ \rho_G \left( J_{\mu}(x', 0), \frac{\partial J_{\mu}}{\partial t}(x, t) \right) \right]. \quad (19)$$

With elementary manipulations this can be brought into the form

$$R_{\mu} = \lim_{\eta \rightarrow 0^+} \frac{m^2}{n^2 e^2 \Omega} \frac{1}{kT} \int_{\eta}^{\infty} dt \int d^3x d^3x' \times \text{Tr} \left( \rho_G \frac{\partial J_{\mu}(x, t)}{\partial t} \frac{\partial J_{\mu}(x', 0)}{\partial t} \right). \quad (20)$$

For a Hamiltonian of the form

$$H_{e1} = \sum_{\lambda} \int d^3x \psi_{\lambda}^{\dagger}(x) \left( \frac{-\hbar^2 \nabla^2}{2m} \right) \psi_{\lambda}(x) + \sum_{\alpha\beta} \int d^3x \psi_{\alpha}^{\dagger}(x) V_{\alpha\beta}(x) \psi_{\beta}(x), \quad (21)$$

one has

$$\frac{\partial J_{\mu}}{\partial t}(\dot{x}, t) = -\frac{1}{m} \sum_{\alpha\beta} \psi_{\beta}^{\dagger}(x, t) \nabla_{\mu} V_{\alpha\beta}(x, t) \psi_{\alpha}(x, t), \quad (22)$$

and therefore

$$R_{\mu} = \lim_{\eta \rightarrow 0^+} \frac{1}{n^2 e^2 \Omega} \frac{1}{kT} \int_{\eta}^{\infty} dt \int d^3x d^3x' \times \text{Tr} \left( \rho_G \sum_{\alpha\beta} \psi_{\beta}^{\dagger}(x, t) \nabla_{\mu} V_{\beta\alpha}(x, t) \psi_{\alpha}(x, t) \times \sum_{\gamma\delta} \psi_{\gamma}^{\dagger}(x', 0) \nabla_{\mu} V_{\gamma\delta}(x', 0) \psi_{\delta}(x', 0) \right). \quad (23)$$

It is this expression which will form the starting point of the perturbative expansion to be developed in the following sections. This expression is somewhat more general than that given by Rousseau, Stoddart, and March<sup>7</sup> in that the Fermi factors have not been explicitly identified. That is in fact possible only if the potential is spin and time independent.

Similar expressions for inverse transport coefficients such as (23) were first given by Kirkwood for the Brownian friction constant<sup>9</sup> and are well known in the theory of transport in liquids.<sup>10</sup> The

recent reintroduction of these formulas into the solid-state literature<sup>6,7</sup> has been criticized by Chassib, Gilbert, and Morgan<sup>11</sup> because it can be argued that when  $\eta$  is set equal to zero in Eq. (23), these expressions are formally equal to zero.<sup>12</sup> Kubo<sup>8</sup> was also well aware of these formal difficulties. On the other hand, perturbative expansions of these expressions give very reasonable results. For example, Eq. (23) yields the Grüneisen formula for the phonon contribution to the resistivity in zeroth order,<sup>12</sup> and, as will be shown in Sec. III, the familiar resistivity formula due to a single moment is also reproduced. However, the question of validity of these expressions have not been satisfactorily resolved. Our speculation is that in view of the above reported successes of Eq. (23), this formula is more generally valid than the formal objections might lead us to suspect.<sup>13</sup>

### III. CONDUCTION-ELECTRON-SPIN INTERACTION

We now consider the calculation of the resistivity of an approximately free-electron metal in which the spins interact with the conduction electrons and with each other. The Hamiltonian is the sum of two parts

$$H = H_{e1} + H_M, \quad (24)$$

where  $H_M$  is the spin-spin interaction responsible for magnetic ordering. We shall take  $H_{e1}$  to be of the free-electron form on the assumption that the disordered arrangement of atomic potentials is not responsible for the resistance minimum. We take the conduction-electron-spin interaction to be of the standard Kondo form

$$V_{\alpha\beta}(x, t) = J \vec{\sigma}_{\alpha\beta} \cdot \sum_{\alpha} \vec{S}_{\alpha}(t) \delta(x - r_{\alpha}), \quad (25)$$

where  $\vec{\sigma}_{\alpha\beta}$  are Pauli spin matrices,  $\vec{S}_{\alpha}(t)$  is the time-dependent spin, and  $r_{\alpha}$  is the position of the  $\alpha$ th spin.

We shall now proceed with a perturbative expansion of the resistivity formula (23) in the conduction electron-spin interaction equation (25).

Let us first develop some notation. The Fourier transforms of the electron fields are defined in terms of creation and annihilation operators according to

$$\psi_{\alpha}^{\dagger}(x, t) = \int \frac{d^3k_1}{(2\pi)^3} e^{-i\mathbf{k}_1 \cdot \mathbf{x}} a_{\mathbf{k}_1, \alpha}^{\dagger}(t), \quad (26)$$

$$\psi_{\beta}(x, t) = \int \frac{d^3k_2}{(2\pi)^3} e^{-i\mathbf{k}_2 \cdot \mathbf{x}} a_{\mathbf{k}_2, \beta}(t).$$

We also define

$$S^i(\mathbf{k}, t) = \sum_{\alpha} e^{-i\mathbf{k} \cdot \mathbf{r}_{\alpha}} S_{\alpha}^i(t). \quad (27)$$

Then from Eqs. (25)–(27),

$$\int d^3x \psi_{\beta}^{\dagger}(x, t) \nabla_{\mu} V_{\beta\alpha}(x, t) \psi_{\alpha}(x, t)$$

$$= J \int \frac{d^3 k_1}{(2\pi)^3} \frac{d^3 k_2}{(2\pi)^3} i(k_1 - k_2)_\mu \times \sum_i a_{k_1\beta}^\dagger(t) \sigma_{\beta\alpha}^i a_{k_2\alpha}(t) S^i(k_1 - k_2, t). \quad (28)$$

Since we are carrying out a perturbative expansion in the conduction-electron-spin interaction, we also have

$$a_{k\beta}^\dagger(t) = a_{k\beta}^\dagger e^{i\epsilon_k t/\hbar}, \quad a_{k\beta}(t) = a_{k\beta} e^{-i\epsilon_k t/\hbar}, \quad (29)$$

with

$$\epsilon_k = \hbar^2 k^2 / 2m.$$

Matrix elements of the form

$$\text{Tr} \{ \hat{\rho}_G a^\dagger a \cdots a^\dagger a \}$$

may be evaluated with the aid of the relations given in the book by Fetter and Walecka.<sup>14</sup>

The  $J^2$  contribution to the resistivity may be evaluated as the zeroth-order perturbative term

$$R_\mu^{(2)} = \frac{1}{e^2 n^2 \Omega} \frac{J^2}{kT} \int_0^\infty dt \int \frac{d^3 k_1}{(2\pi)^3} \frac{d^3 k_2}{(2\pi)^3} \frac{d^3 k_3}{(2\pi)^3} \frac{d^3 k_4}{(2\pi)^3} i(k_1 - k_2)_\mu i(k_3 - k_4)_\mu \times \sum_{\alpha\beta\gamma\delta} \sum_{ij} \sigma_{\beta\alpha}^i \sigma_{\gamma\delta}^j e^{-\epsilon t} e^{i(\epsilon_{k_1} - \epsilon_{k_2})t/\hbar} \text{Tr}[\rho_G^0 S^i(k_1 - k_2, t) S^j(k_3 - k_4, 0)] \times (2\pi)^3 \delta(k_1 - k_4) \delta_{\beta\delta} (2\pi)^3 \delta(k_2 - k_3) \delta_{\alpha\gamma} n_1^0 (1 - n_3^0), \quad (30)$$

where  $n_1^0$  is the Fermi function corresponding to momentum 1, etc., and  $\rho_G^0$  is the grand canonical statistical operator in the absence of a conduction-electron-spin interaction. Defining a second-order spin correlation function

$$S^{(2)}(k, \omega) \equiv \sum_i \int dt e^{i\omega t} \text{Tr}[\rho_G^0 S^i(k, t) S^i(-k, 0)], \quad (31)$$

we find that Eq. (30) may be simplified to

$$R_\mu^{(2)} = \frac{2J^2}{e^2 n^2 \Omega kT} \int \frac{d\omega}{2\pi} \int \frac{d^3 k_1}{(2\pi)^3} \frac{d^3 k_3}{(2\pi)^3} \times \frac{(k_1 - k_3)_\mu (k_3 - k_1)_\mu}{i(\epsilon_{k_1} - \epsilon_{k_3} - \hbar\omega)/\hbar - \epsilon}$$

$$\times S^{(2)}(k_1 - k_3, \omega) n_1^0 (1 - n_3^0). \quad (32)$$

Thus, the second-order contribution to the resistivity is determined by a second-order spin-correlation function. More generally, one finds that the  $n$ th-order contribution to the resistivity is determined by an  $n$ th-order spin-correlation function.

The contributions responsible for the resistance minimum occur in third and higher orders. In evaluating the  $J^3$  contribution one must keep in mind the cancellation of disconnected parts and remove such terms as they appear. Thus

$$R_\mu^{(3)} = \frac{1}{e^2 n^2 \Omega} \frac{1}{kT} \int_0^\infty dt \int d^3 x \int d^3 x' \int_{-\infty}^\infty dt_1 \int d^3 x_1 \left( \frac{-i}{\hbar} \right) \text{Tr} \left[ \rho_G^0 T \left( \sum_{\alpha\beta} \psi_\beta^\dagger(x, t) \nabla_\mu V_{\beta\alpha}(x, t) \psi_\alpha(x, t) \times \sum_{\gamma\delta} \psi_\gamma^\dagger(x', 0) \nabla_\mu V_{\gamma\delta}(x', 0) \psi_\delta(x', 0) \sum_{\epsilon\phi} \psi_\epsilon^\dagger(x_1, t_1) V_{\epsilon\phi}(x_1, t_1) \psi_\phi(x_1, t_1) \right) \right]_{\text{connected}}, \quad (33)$$

where the symbol  $T$  means that the operators inside the trace should be time ordered.

In momentum space,

$$R_\mu^{(3)} = \frac{1}{e^2 n^2 \Omega} \frac{J^3}{kT} - \left( \frac{i}{\hbar} \right) \int_0^\infty dt e^{-\epsilon t} \int_{-\infty}^\infty dt' e^{-\epsilon|t'|} \int \frac{d^3 k_1}{(2\pi)^3} \frac{d^3 k_2}{(2\pi)^3} \frac{d^3 k_3}{(2\pi)^3} \frac{d^3 k_4}{(2\pi)^3} \frac{d^3 k_5}{(2\pi)^3} \frac{d^3 k_6}{(2\pi)^3} i(k_1 - k_2)_\mu i(k_3 - k_4)_\mu \times \sum_{\alpha\beta\gamma\delta\epsilon\phi} \text{Tr} \left\{ \rho_G^0 T \left[ \left( \sum_i a_{k_1\beta}^\dagger(t) \sigma_{\beta\alpha}^i a_{k_2\alpha}(t) S^i(k_1 - k_2, t) \right) \left( \sum_j a_{k_3\gamma}^\dagger(0) \sigma_{\gamma\delta}^j a_{k_4\delta}(0) S^j(k_3 - k_4, 0) \right) \left( \sum_k a_{k_5\epsilon}^\dagger(t') \sigma_{\epsilon\phi}^k a_{k_6\phi}(t') S^k(k_5 - k_6, t') \right) \right] \right\}_{\text{connected}}. \quad (34)$$

The subscript "connected" now means that in the reduction of this expression all terms with  $k_1 = k_2$ ,  $k_3 = k_4$ , or  $k_5 = k_6$  are to be removed.

The evaluation of expression (34) involves a lengthy but straightforward calculation. To express the result compactly let us first consider a

triple-spin-correlation function in some detail. We define

$$-i\epsilon^{ijk} \text{Tr}[\rho_G^0 S^i(k_1, t_1) S^j(k_2, t_2) S^k(k_3, t_3)] \equiv \int \frac{d\omega_1}{(2\pi)} \frac{d\omega_3}{(2\pi)} e^{-i\omega_1(t_1 - t_2)}$$

$$\times e^{i\omega_3(t_3-t_2)} S^{(3)}(k_1, k_3, \omega_1, \omega_3), \quad (35)$$

where two momentum labels are sufficient since the only correlation functions to be considered are those for which  $k_1 + k_2 + k_3 = 0$ . The functions  $S^{(3)}(k_1, k_3, \omega_1, \omega_3)$  can be shown to satisfy

$$[S^{(3)}(k_1, k_3, \omega_1, \omega_3)]^\dagger = S^{(3)}(-k_3, -k_1, \omega_3, \omega_1). \quad (36)$$

It is important to remark that  $S^{(2)}(k, \omega)$  and  $S^{(3)}(k_1, k_3, \omega_1, \omega_3)$  are not independent. They are related by sum rules of which the most important for our considerations are

$$\begin{aligned} R_\mu^{(3)} = & \frac{2J^3}{e^2 n^2 k T \Omega} \left( \frac{-i}{\hbar} \right) \int \frac{d^3 k_1}{(2\pi)^3} \frac{d^3 k_3}{(2\pi)^3} \frac{d^3 k_5}{(2\pi)^3} \int \frac{d\omega_1}{2\pi} \frac{d\omega_3}{2\pi} [i(k_1 - k_3)_\mu i(k_3 - k_1)_\mu + i(k_3 - k_5)_\mu i(k_5 - k_1)_\mu] \\ & \times S^{(3)}(k_1 - k_5, k_5 - k_3, \omega_1, \omega_3) \left( \frac{(-1)n_1^0(1-n_5^0)n_3^0}{[i(\epsilon_1 - \epsilon_5 - \hbar\omega_1)/\hbar - \epsilon][i(\epsilon_5 - \epsilon_3 + \hbar\omega_3)/\hbar + \epsilon]} \right. \\ & \left. + \frac{(-1)n_5^0(1-n_1^0)(1-n_3^0)}{[i(\epsilon_5 - \epsilon_1 - \hbar\omega_1)/\hbar - \epsilon][i(\epsilon_3 - \epsilon_5 + \hbar\omega_3)/\hbar + \epsilon]} \right). \end{aligned} \quad (39)$$

This equation is the principal result of this section. It expresses the relation between the third-order contribution to the resistivity and the triple-spin-correlation function. In performing the momentum integrations a cutoff must be introduced to avoid ultraviolet divergences. This divergence is due to our neglect of the dependence on momentum transfer of the conduction-electron-spin interaction. As discussed by Kondo, this divergence is not relevant to the resistance-minimum phenomenon.<sup>15</sup>

It is instructive to examine how this expression gives rise to a resistance minimum in the classic Kondo problem, a single local moment interacting with the conduction electrons. In this case the magnetic ordering Hamiltonian  $H_M$  in Eq. (24) is equal to zero. Since

$$S^i(k, t) \equiv e^{iH_M t/\hbar} \left( \sum_a e^{-i\mathbf{k} \cdot \mathbf{r}_a} S_a^i(0) \right) e^{-iH_M t/\hbar}, \quad (40)$$

one has, in the case of a single spin,

$$S^i(k, t) = e^{-i\mathbf{k} \cdot \mathbf{r}_a} S_a^i. \quad (41)$$

Then in Eq. (35) one has

$$-i\epsilon^{ijk} \text{Tr}[\rho_G^0 S^i(k_1, t_1) S^j(k_2, t_2) S^k(k_3, t_3)] = S(S+1), \quad (42)$$

when  $k_1 + k_2 + k_3 = 0$  and  $S$  is the spin of the local moment. Therefore,

$$S^{(3)}(k_1, k_3, \omega_1, \omega_3) = S(S+1)(2\pi)\delta(\omega_1)(2\pi)\delta(\omega_3) \quad (43)$$

in the single-local-moment problem.

If one combines Eqs. (39) and (43) and performs the angular integrations, one finds

$$\int \frac{d\omega_1}{2\pi} S^{(3)}(k_1, k_3, \omega_1, \omega_3) = S^{(2)}(-k_3, \omega_3) \quad (37)$$

and

$$\int \frac{d\omega_3}{2\pi} S^{(3)}(k_1, k_3, \omega_1, \omega_3) = S^{(2)}(k_1, \omega_1). \quad (38)$$

Thus any statement we make regarding triple-spin-correlation functions implies a statement about the more experimentally accessible double-spin-correlation function.

Then the final expression for the third-order contribution to the resistivity may be written

$$\begin{aligned} R_\mu^{(3)} = & \frac{2S(S+1)}{3e^2 n^2 \Omega} \frac{J^3(-i/\hbar)}{kT(2\pi)^9} \\ & \times \int k_1^2 dk_1 k_3^2 dk_3 k_5^2 dk_5 (k_1^2 + k_3^2 + k_5^2) \\ & \times \left( \frac{(-1)n_1^0(1-n_5^0)n_3^0}{[i(\epsilon_1 - \epsilon_5)/\hbar - \epsilon][i(\epsilon_5 - \epsilon_3)/\hbar + \epsilon]} \right. \\ & \left. + \frac{(-1)n_5^0(1-n_1^0)(1-n_3^0)}{[i(\epsilon_5 - \epsilon_1)/\hbar - \epsilon][i(\epsilon_3 - \epsilon_5)/\hbar + \epsilon]} \right). \end{aligned} \quad (44)$$

We now use the general relation that for a function  $f(k_1)$  invariant under  $k_1 \rightarrow -k_1$ , and  $k_5 > 0$ ,

$$\int_0^\infty \frac{dk_1 f(k_1)}{i(\epsilon_1 - \epsilon_5)/\hbar - \epsilon} = \int_0^\infty \frac{dk_1 f(k_1)}{i(\epsilon_5 - \epsilon_1)/\hbar - \epsilon} = -\frac{\pi m f(k_5)}{\hbar k_5}. \quad (45)$$

Then

$$\begin{aligned} R_\mu^{(3)} = & \frac{-2S(S+1)J^3\pi m}{3e^2 n^2 \Omega (2\pi)^9 \hbar^2 kT} \int k_5^3 dk_5 k_3^2 dk_3 \\ & \times (2k_5^2 + k_3^2) n_5^0 (1 - n_5^0) \frac{2n_3^0 - 1}{\epsilon_5 - \epsilon_3}. \end{aligned} \quad (46)$$

Although it is possible, in principle, to evaluate this expression exactly if a suitable ultraviolet cutoff is introduced, to demonstrate a resistance minimum it is sufficient to consider only the qualitative character of this expression in the low-temperature limit.

First consider a general integral of the type

$$I(T) = \frac{1}{kT} \int d\epsilon n^0(1-n^0)f(\epsilon), \quad (47)$$

where  $f(\epsilon)$  is a smoothly varying function of  $\epsilon$ . At low temperatures the limits in the integral can be extended to  $-\infty$  and  $+\infty$  without appreciable error. Then

$$I(T) = \int_{-\infty}^{\infty} dx \frac{1}{e^x + 1} \frac{1}{1 + e^{-x}} f(kTx + \mu), \quad (48)$$

where  $\mu$  is the Fermi energy. The significant contributions to the integral come from  $-1 < x < 1$ , so that for low temperatures,

$$I(T) \cong f(\mu). \quad (49)$$

If one combines this result with Eq. (46), the third-order contribution to the resistivity is proportional in the low-temperature limit to an integral of the form

$$R_{\mu}^{(3)} \propto -J^3 \int d\epsilon_3 \frac{(2n_3^0 - 1)g(\epsilon_3)}{\mu - \epsilon_3}, \quad (50)$$

where  $g(\epsilon_3)$  is a positive smoothly varying function of  $\epsilon_3$ . This is exactly the form of the resistivity obtained by Kondo in his third-order calculation. It is easily shown that

$$R_{\mu}^{(3)} \propto +J^3 \ln kT. \quad (51)$$

When this is added to the phonon contribution which decreases with decreasing temperature and  $J$  is negative (antiferromagnetic) the result is a minimum in the resistance as discussed by Kondo.<sup>15</sup> The divergence at  $T = 0^\circ\text{K}$  is removed by the higher-order terms in the perturbative expansion of the resistivity.

#### IV. RESISTANCE MINIMA IN AMORPHOUS FERROMAGNETS

The neglect of the interaction between spins is clearly not justifiable in alloys with appreciable concentrations of ferromagnetic elements and which tend to magnetically order as the temperature is lowered. As shown in Sec. III, a perturbative calculation of the resistivity due to the conduction-electron-spin interaction will in the general case have the  $n$ th-order contribution determined by an  $n$ th-order spin-correlation function.

In this section we address the question of what properties the spin correlation functions must have in order that the sum of the spin and phonon contributions yield a minimum in the resistance at some temperature. We will assume that the Kondo mechanism continues to hold, i.e., in third order the influence of the Fermi distribution gives rise to a  $\ln kT$  term in the resistivity. The divergence at zero temperature is presumably canceled by the higher-order contributions, as in the usual theories of the Kondo effect in dilute alloys. Specifically, we assume that, in third order, one may write

$$R_{\mu}^{(3)} \cong A(T) \ln kT + B(T), \quad (52)$$

where  $A(T)$  and  $B(T)$  are both well behaved in the low-temperature limit. We will calculate the coefficient  $A(T)$  and determine what a nonzero  $A(T)$  implies about the spin correlation functions. This in turn can be related to a physical statement about the nature of the spin excitation spectrum in amorphous ferromagnetic alloys with resistance minima.

We begin the calculation by establishing when the triple-spin-correlation functions are zero. From Eq. (35) one may readily derive

$$\begin{aligned} S^{(3)}(k_1, k_3, \omega_1, \omega_3) &= -i\epsilon^{ijk} \sum_{lmn} e^{\beta\Omega} e^{-\beta E_l} \langle l | S^i(k_1, 0) | m \rangle \\ &\times \langle m | S^j(k_2, 0) | n \rangle \langle n | S^k(k_3, 0) | l \rangle \\ &\times (2\pi) \delta\left(\omega_1 - \frac{E_m - E_l}{\hbar}\right) (2\pi) \delta\left(-\omega_3 + \frac{E_n - E_l}{\hbar}\right). \end{aligned} \quad (53)$$

The low-temperature limit of this expression is

$$\begin{aligned} \lim_{\beta \rightarrow \infty} S^{(3)}(k_1, k_3, \omega_1, \omega_3) &= -i\epsilon^{ijk} \sum_{mn} \langle 0 | S^i(k_1, 0) | m \rangle \\ &\times \langle m | S^j(k_2, 0) | n \rangle \langle n | S^k(k_3, 0) | 0 \rangle \\ &\times (2\pi) \delta\left(\omega_1 - \frac{E_m - E_0}{\hbar}\right) (2\pi) \delta\left(-\omega_3 + \frac{E_n - E_0}{\hbar}\right). \end{aligned} \quad (54)$$

Clearly, since  $E_m \geq E_0$  and  $E_n \geq E_0$ ,

$$\begin{aligned} \lim_{\beta \rightarrow \infty} S^{(3)}(k_1, k_3, \omega_1, \omega_3) &= \text{finite} \quad \omega_1, \omega_3 \geq 0 \\ &= 0 \quad \text{otherwise}. \end{aligned} \quad (55)$$

It is possible to make a statement regarding the manner in which  $S^{(3)}(k_1, k_3, \omega_1, \omega_3)$  approaches zero for  $\omega_1$  or  $\omega_3 < 0$ . It may be readily proved from Eq. (53) that

$$\begin{aligned} S^{(3)}(k_1, k_3, \omega_1, \omega_3) &= e^{\beta\hbar\omega_3} S^{(3)}(k_3, k_2, -\omega_3, \omega_1 - \omega_3) \\ &= e^{\beta\hbar\omega_1} S^{(3)}(k_2, k_1, \omega_3 - \omega_1, -\omega_1). \end{aligned} \quad (56)$$

Hence if  $\omega_1 < 0$  but  $\omega_3 \geq 0$ ,  $S^{(3)}(k_1, k_3, \omega_1, \omega_3)$  goes to zero as does  $e^{\beta\hbar\omega_1}$  times a function which is finite in the limit  $\beta \rightarrow \infty$ .

To simplify the expression for the resistivity let us define

$$\begin{aligned} f_{\mu}(\epsilon_1, \epsilon_3, \epsilon_5, \omega_1, \omega_3) &\equiv \frac{(k_1^2)^{1/2}}{(2\pi)^9} \\ &\times \int d\Omega_1 d\Omega_3 d\Omega_5 [i(k_1 - k_3)_{\mu} i(k_3 - k_1)_{\mu} \\ &+ i(k_3 - k_5)_{\mu} i(k_5 - k_1)_{\mu}] \\ &\times S^{(3)}(k_1 - k_5, k_5 - k_3, \omega_1, \omega_3), \end{aligned} \quad (57)$$

where the  $d\Omega_i$  are angular integrations in  $k$  space. We argue that  $f_{\mu}$  should vary smoothly for  $\epsilon_1, \epsilon_3$ ,

or  $\epsilon_5$  in the vicinity of the chemical potential  $\mu$ . This is inherent in our perturbative treatment of the conduction electron-spin interaction, a treatment based on the assumption that the conduction

electron-spin interaction only weakly influences the interaction between spins.

Then the expression for the resistivity obtained by combining Eqs. (39), (45), and (57) is

$$R_{\mu}^{(3)} = \frac{2J^3(-i)(-\pi m)}{e^2 n^2 \Omega k T \hbar^2} \int k_3^2 dk_3 \int k_5^2 dk_5 \int \frac{d\omega_1}{2\pi} \frac{d\omega_3}{2\pi} \\ \times \left( \frac{(-1)n^0(\epsilon_5 + \hbar\omega_1)[1 - n^0(\epsilon_5)]n^0(\epsilon_3)}{i(\epsilon_5 - \epsilon_3 + \hbar\omega_3)/\hbar + \epsilon} f_{\mu}(\epsilon_5 + \hbar\omega_1, \epsilon_3, \epsilon_5, \omega_1, \omega_3) \right. \\ \left. + \frac{(-1)n^0(\epsilon_3)[1 - n^0(\epsilon_5 - \hbar\omega_1)][1 - n^0(\epsilon_3)]}{i(\epsilon_3 - \epsilon_5 + \hbar\omega_3)/\hbar + \epsilon} f_{\mu}(\epsilon_5 - \hbar\omega_1, \epsilon_3, \epsilon_5, \omega_1, \omega_3) \right), \quad (58)$$

where we have now written explicitly the argument of the Fermi function  $n^0(\epsilon)$ . As stated in Sec. III, an ultraviolet cutoff must be introduced on the momentum integrals to account crudely for the neglect of the dependence on momentum transfer of the conduction-electron-spin interaction.

Let us discuss the  $d\epsilon_3 d\omega_3$  integral first. We write

$$n^0(\epsilon_3) = \frac{1}{2}[2n^0(\epsilon_3) - 1] + \frac{1}{2}, \quad 1 - n^0(\epsilon_3) = -\frac{1}{2}[2n^0(\epsilon_3) - 1] + \frac{1}{2}. \quad (59)$$

Only the  $2n^0(\epsilon_3) - 1$  terms can give rise to a contribution proportional to  $\ln kT$ , and therefore we shall consider only these terms. Then it is readily seen that the  $d\epsilon_3 d\omega_3$  integral is of the general form

$$I(T) \cong \int_0^c d\epsilon \int_{-\infty}^{\infty} dx \frac{[2n^0(\epsilon) - 1]f(\epsilon, x)}{x - \epsilon}, \quad (60)$$

where  $f(\epsilon, x)$  is a smoothly varying function of  $\epsilon$  for  $\epsilon$  near the chemical potential  $\mu$ , and  $c$  is an ultraviolet cutoff with  $c \gg \mu$ . We approximate  $2n^0(\epsilon) - 1$  by

$$\begin{aligned} 2n^0(\epsilon) - 1 &\cong +1, & \epsilon - \mu < 2/\beta, \\ &\cong -\frac{1}{2}\beta(\epsilon - \mu), & -2/\beta < \epsilon - \mu < 2/\beta, \\ &\cong -1, & \epsilon - \mu > 2/\beta. \end{aligned} \quad (61)$$

For the purpose of evaluating this integral in the low-temperature limit let us choose two energies  $L$  and  $U$  such that

$$0 < L < \mu - 2/\beta \quad \text{and} \quad \mu + 2/\beta < U < c. \quad (62)$$

Then we break up the integral as follows:

$$I(T) = \int_{-\infty}^{\infty} dx \left( \int_0^L d\epsilon \frac{f(\epsilon, x)}{x - \epsilon} + \int_L^{\mu - 2/\beta} d\epsilon \frac{f(\epsilon, x)}{x - \epsilon} + \int_{\mu - 2/\beta}^{\mu + 2/\beta} d\epsilon \frac{1}{2}\beta(\mu - \epsilon) \frac{f(\epsilon, x)}{x - \epsilon} \right. \\ \left. + \int_{\mu + 2/\beta}^U d\epsilon \frac{f(\epsilon, x)}{\epsilon - x} + \int_U^c d\epsilon \frac{f(\epsilon, x)}{\epsilon - x} \right). \quad (63)$$

In the limit of low temperatures ( $\beta \rightarrow \infty$ ), we can choose  $L$  and  $U$  sufficiently close to  $\mu$  such that  $f(\epsilon, x)$  does not vary appreciably in the range  $L < \epsilon < U$ . Hence, we have approximately in the limit of low temperatures that  $I(T)$  consists of three terms

$$I(T) = \int_{-\infty}^{\infty} dx \left[ \int_0^L d\epsilon \frac{f(\epsilon, x)}{x - \epsilon} + \int_U^c d\epsilon \frac{f(\epsilon, x)}{\epsilon - x} + f(\mu, x) \left( \int_L^{\mu - 2/\beta} \frac{d\epsilon}{x - \epsilon} - \int_{\mu + 2/\beta}^U \frac{d\epsilon}{\epsilon - x} \right) \right], \quad (64)$$

where

$$\left( \int \right) = -[+1 - \frac{1}{2}\beta(\mu - x)] \ln |x - \mu + (2/\\beta)| - [+1 + \frac{1}{2}\beta(\mu - x)] \ln |x - \mu - 2/\beta| + \ln |x - L| - \ln |U - x| - 2.$$

Since in our case  $f(\epsilon, x)$  is not expected to change sign as  $\epsilon$  goes from  $x - \eta$  to  $x + \eta$ , the first two terms in Eq. (64) will be finite, and the endpoints of the integrations will cancel the  $\ln|x - L||U - x|$  contribution to the third term. The contribution to the resistivity proportional to  $\ln kT$  arises from the remainder of the third term in Eq. (64) for the  $x$  integral in the range

$$2/\beta + \mu > x > -2/\beta + \mu.$$

Thus,  $I(T)$  is given approximately by

$$I(T) = -2 \ln kT \int_{\mu-2/\beta}^{\mu+2/\beta} dx f(\mu, x) + B(T), \quad (65)$$

where  $B(T)$  is well behaved in the low-temperature limit.

Combining Eqs. (58), (60), and (65) the  $\ln kT$  term in the third-order contribution to the resistivity is approximately

$$R_\mu^{(3)} \cong + \frac{2 \ln kT J^3 \pi m}{e^2 n^2 \Omega k T \hbar^2} \int k_5^2 dk_5 \int \frac{d\omega_1}{2\pi} x \left[ \right], \quad (66)$$

where

$$\left[ \right] = \int_{\mu-\epsilon_5-2/\beta}^{\mu-\epsilon_5+2/\beta} \frac{d\hbar\omega_3}{2\pi} n^0(\epsilon_5 + \hbar\omega_1) [1 - n^0(\epsilon_5)] f_\mu(\epsilon_5 + \hbar\omega_1, \mu, \epsilon_5; \omega_1, \omega_3) \\ + \int_{\epsilon_5-\mu-2/\beta}^{\epsilon_5-\mu+2/\beta} \frac{d\hbar\omega_3}{2\pi} n^0(\epsilon_5) [1 - n^0(\epsilon_5 - \hbar\omega_1)] f_\mu(\epsilon_5 - \hbar\omega_1, \mu, \epsilon_5; \omega_1, \omega_3).$$

We now want to consider the coefficient of  $\ln kT$  in the low-temperature limit. Clearly, the endpoints of the  $d(\hbar\omega_3)$  integrals approach each other as  $\beta \rightarrow \infty$ . The only way for the coefficient of  $\ln kT$  to remain nonzero is for  $f_\mu$  to contain a term with a  $\delta$ -function dependence on  $\hbar\omega_3$ , i.e.,

$$f_\mu(\epsilon_5 + \hbar\omega_1, \mu, \epsilon_5; \omega_1, \omega_3) = g_\mu^{(1)}(\epsilon_5, \omega_1) \\ \times \delta(\hbar\omega_3 + \epsilon_5 - \mu) + \text{analytic in } \omega_3 \quad (67)$$

and

$$f_\mu(\epsilon_5 - \hbar\omega_1, \mu; \epsilon_5, \omega_1, \omega_3) = g_\mu^{(2)}(\epsilon_5, \omega_1) \\ \times \delta(\hbar\omega_3 - \epsilon_5 + \mu) + \text{analytic in } \omega_3.$$

From Eqs. (55) and definition (57) we have that in the low-temperature limit the  $f_\mu$  are nonzero only for  $\omega_1, \omega_3 \geq 0$ . In the first term in (66) we must have  $\epsilon_5 > \mu$  and  $\epsilon_5 + \hbar\omega_1 < 0$ , and in the second term in (66) we must have  $\epsilon_5 - \hbar\omega_1 > \mu$ ,  $\epsilon_5 < \mu$  due to the Fermi factors  $n^0$ . Combining all these statements we find that the coefficient of the  $\ln kT$  can remain nonzero only if  $f_\mu$  has a  $\delta$ -function dependence on  $\omega_1$  and  $\omega_3$  as follows:

$$f_\mu(\epsilon_1, \epsilon_3, \epsilon_5; \omega_1, \omega_3) = g_\mu(\epsilon_1, \epsilon_3, \epsilon_5) \delta(\omega_1) \delta(\omega_3) \\ + \text{analytic in } \omega_1 \text{ and } \omega_3. \quad (68)$$

Because of the definition of  $f_\mu$  [Eq. (57)], we find that the only way for the coefficient of the  $\ln kT$  term in the third-order contribution to the resistivity to remain nonzero in the low-temperature limit is for the triple-spin-correlation function to

have the general behavior

$$S^{(3)}(k_1, k_3, \omega_1, \omega_3) = S(k_1, k_3) \delta(\omega_1) \delta(\omega_3) \\ + \text{analytic in } \omega_1 \text{ and } \omega_3. \quad (69)$$

This conclusion appears unavoidable. Because of the sum rules (37) and (38) we must also have

$$S^{(2)}(k, \omega) = S(k) \delta(\omega) + \text{analytic in } \omega. \quad (70)$$

A physical statement of Eq. (70) is that the alloy must have a finite density of zero-frequency excitations in order that the coefficient of the  $\ln kT$  term remain nonzero in the low-temperature limit.

Our considerations must be qualified somewhat when the contributions of higher-order terms in the resistivity are included. When these terms become comparable to the third order term (for temperatures at and below the Kondo temperature) no statement can be made. Hence, rigorously speaking we can only prove that the observation of a resistance minimum implies a finite density of states for frequencies in the range  $-kT_K \lesssim \hbar\omega \lesssim kT_K$ . A safe rewording of our statement appears to be that the observation of a resistance minimum implies a finite density of near-zero-frequency excitations.

## V. CONCLUSION

To summarize, we have considered the problem of resistivity minima in alloys with non-negligible spin-spin interactions. The resistivity due to the conduction electron-spin interaction is evaluated using expressions for inverse transport coefficient



cients. In a perturbative expansion it is found that the  $n$ th-order term is determined by an  $n$ th-order spin-correlation function. The coefficient of  $\ln kT$  in the third-order term is found to receive contributions only from the near-zero-frequency parts of the spin-correlation functions in the low-temperature limit.

Thus, unless some other mechanism than the conduction electron spin interaction is operating, it appears that the spin excitation spectra of amorphous ferromagnets exhibiting resistivity minima must have a finite density of near-zero-frequency excitations ( $\hbar\omega < kT_F$ ). This can, in principle, be verified by neutron scattering if the experimental difficulties in obtaining sufficient amorphous material can be overcome.

Without favoring any particular viewpoint concerning the origin of the near zero frequency excitations, we would like to mention two possibilities that have been discussed in the literature. Tsuei has suggested that because of the statistical distribution of the moments in amorphous alloys a

certain fraction of the spins may be only weakly coupled to the other spins and therefore relatively little energy may be required for a spin flip. This view is supported by Mössbauer studies which indicate that a non-negligible fraction of the iron atoms in these alloys have near-zero hyperfine field.<sup>16</sup> Madhukar and Hasegawa have emphasized that the amorphous nature of these alloys may lead to only short-range magnetic order. There would consequently be low- $\omega$  magnons localized in real space and therefore of high  $q$ .<sup>4</sup> Other models for the spin excitation spectra could be tested directly by substituting into Eq. (39).

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